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For: METHOD FOR PRODUCING AROMATIC COMPOUND ISOMERS

Also enclosed are:

- ☒ 7 Sheets of drawings
☒ Recordation Form Cover Sheet - Patents Only and an Assignment of the invention to Toray Industries, Inc.
☒ Claim for Priority Under 35 U.S.C. §119 with certified copy of JP 11-240393
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METHOD FOR PRODUCING AROMATIC COMPOUND ISOMERS

BACKGROUND OF THE INVENTION

1. Field of the Invention:

The present invention relates to a method for producing aromatic compound isomers. More precisely, the invention relates to a method of adsorptive separation for producing intended aromatic compound isomers by the use of a zeolite-containing adsorbent and a desorbent.

2. Description of the Related Art:

Aromatic compounds are useful for starting materials for synthetic products and for intermediates for medicines and agricultural chemicals. For obtaining a specific isomer of an aromatic compound, known is a method of adsorptive separation that comprises contacting an isomer mixture with a zeolite-containing adsorbent.

For example, Japanese Patent Publication No. 45457/1989 discloses a method of using an adsorbent of faujasite zeolite for adsorptive separation of 2,6-dichlorotoluene; Japanese Patent Laid-Open No. 330025/1992 discloses a method of using an adsorbent of faujasite zeolite that indispensably contains potassium and lead ions, for adsorptive separation of m-dichlorobenzene; and Japanese Patent Publication No. 46933/1992 discloses a

process of isomerizing halogenobenzene derivatives, in which HCl formed in the step of isomerization is first removed through distillation or the like, and a specific isomer is thereafter separated from the isomer mixture through adsorptive separation by the use of a zeolite adsorbent.

Regarding heterocyclic compounds, Japanese Patent Laid-Open Nos. 143162/1997 and 251230/1998 and EP 173,440 disclose a method of using an adsorbent of faujasite zeolite for adsorptive separation of a compound having a pyridine ring structure.

In the process of adsorptive separation of aromatic compound isomers by the use of a zeolite-containing adsorbent, the adsorbent is degraded while used continuously. In the process, therefore, the adsorbent used must be exchanged for a fresh one in good time or must be regenerated, for example, by burning it. Accordingly, a technique for prolonging the life or the regeneration cycle of adsorbent, if possible, will produce great industrial merits.

In the process of the above-mentioned Japanese Patent Publication No. 46933/1992 for adsorptive separation of an aromatic compound isomer by the use of a zeolite adsorbent, the step of removing the side product, HCl from the isomer mixture prior to the next step of

separating the intended isomer by the use of the adsorbent is for preventing the adsorbent used from being degraded. However, the process is still not enough for satisfactorily prolonging the life and the regeneration cycle of the adsorbent to the industrial level.

In ordinary adsorptive separation of aromatic compound isomers, the desorbent used is generally circulated and reused. In the line, however, the desorbent circulated for reuse is often contaminated with minor impurities having existed in the starting aromatic compound isomers and with other impurities having formed through decomposition or reaction of the aromatic compound isomers and/or the desorbent, as the impurities accumulate in the desorbent being used and circulated. As a result, the adsorbent being thus used in the line will be gradually degraded.

SUMMARY OF THE INVENTION

The object of the invention is to efficiently produce aromatic compound isomers through adsorptive separation by the use of a zeolite-containing adsorbent, for which the adsorbent used is prevented from being degraded and its life is prolonged.

To attain its object, the invention provides a method for producing an aromatic compound isomer

substituted with alkyl group(s) and/or halogen atom(s), through adsorptive separation by the use of a zeolite-containing adsorbent and a desorbent, wherein the desorbent is, after having been processed for removing impurities from it, supplied to the adsorptive separation step.

Preferred embodiments of the method of the invention are as follows:

The aromatic compound has a benzene ring or heterocyclic ring structure;

The aromatic compound is ring-substituted with at least one halogen element;

The impurities in the desorbent are removed through any of distillation, desorption or adsorption to a solid adsorbent;

The impurities in the desorbent are removed by replacing a part of the used desorbent with an impurity-free fresh desorbent;

All or part of the desorbent to be supplied to the adsorptive separation step is first continuously or intermittently supplied to a step of removing impurities from it, and then supplied to the adsorptive separation step.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a flow chart for removing impurities from the desorbent in the simulated moving bed process of Example 1.

Fig. 2 is a graph of adsorption recovery versus running time in Example 1 and Comparative Examples 1 and 2.

Fig. 3 is a chart of high-boiling-point impurities accumulated in the desorbent in Example 1.

Fig. 4 is a flow chart for removing impurities from the desorbent in the simulated moving bed process of Example 2.

Fig. 5 is a graph of adsorption recovery versus running time in Example 2 and Comparative Examples 3 and 4.

Fig. 6 is a chart of high-boiling-point impurities in the distillate from the distillation unit and the bottom liquid remained in the unit in Example 2.

Fig. 7 is a graph of adsorption recovery versus running time in Example 3 and Comparative Example 5.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The aromatic compound to be subjected to adsorptive separation according to the invention is an aromatic compound substituted with alkyl group(a) and/or halogen atom(s). The aromatic compound includes a plurality of isomers. In case where the boiling point difference between these isomers is small, the isomers are difficult

to separate from each other through distillation. For the isomers, therefore, suitable is a method of separating them through adsorptive separation.

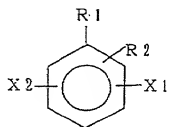
The aromatic compound substituted with alkyl group(s) and/or halogen atom(s) to be processed according to the invention shall have at least one substituent of alkyl groups or halogen atoms, but may optionally have, in addition to it, any other substituent(s), for example, oxygen-containing substituents such as hydroxyl groups, aldehyde groups, carboxyl groups, etc., and nitrogen-containing substituents such as $-\text{NO}_2$, $-\text{NH}_2$, $-\text{NHR}$, $-\text{NR}_2$, $-\text{NRR}'$ (where R and R' are at least one of methyl, ethyl and propyl groups), etc.

The aromatic compound referred to herein is not limited to benzene derivatives only, but includes any others, for example, polycyclic aromatic compounds such as naphthalene, anthracene and phenanthrene compounds, etc., heterocyclic compounds such as pyridine and triazine compounds, etc.

Specific examples of the heterocyclic compounds are α -picoline, β -picoline, γ -picoline, 3,5-lutidine, 2,6-lutidine, 2,4-lutidine, 2,5-lutidine, 3,4-lutidine, 2,3-lutidine, 2,4,6-collidine, 2-ethylpyridine, 3-ethylpyridine, 4-ethylpyridine, 2-vinylpyridine, 3-vinylpyridine, 4-vinylpyridine, 2-pyridinemethanol, 3-

pyridinemethanol, 4-pyridinemethanol, 2-aminopyridine, 3-aminopyridine, 4-aminopyridine, 2-dimethylaminopyridine, 3-dimethylaminopyridine, 4-dimethylaminopyridine, 2-aldehydopyridine, 3-aldehydopyridine, 4-aldehydopyridine, nicotinic acid, isonicotinic acid, picolinic acid, 2-chloropyridine, 3-chloropyridine, 4-chloropyridine, 3,5-dichloropyridine, 2,6-dichloropyridine, 2,4-dichloropyridine, 2,5-dichloropyridine, 3,4-dichloropyridine, 2,3-dichloropyridine, 2-cyanopyridine, 3-cyanopyridine, 4-cyanopyridine, 2-chloro-5-methylpyridine, 2-chloro-4-methylpyridine, 2-chloro-3-methylpyridine, 2-chloro-6-methylpyridine, methylpyrrole, ethylpyrrole, propylpyrrole, dimethylpyrrole, chloropyrrole, methylfuran, ethylfuran, propylfuran, dimethylfuran, chlorofuran, methylthiophene, ethylthiophene, propylthiophene, dimethylthiophene, chlorothiophene, methylquinoline, ethylquinoline, propylquinoline, dimethylquinoline, chloroquinoline, etc.

For the method of the invention, preferred are aromatic compounds substituted with alkyl group(s) and/or halogen atom(s) of the following general formula (I):



..... (I)

wherein R1 and R2 each represent any of halogen atoms or alkyl groups; and X1 and X2 each represent any of halogen atoms, hydrogen atoms or alkyl groups.

More preferred are those that are ring-substituted with at least one halogen element. Especially preferably, the halogen element is chlorine or bromine element. Even more preferred are aromatic compounds that are ring-substituted with chlorine atom(s).

Specific examples of the aromatic compounds (isomers) of formula (I) include xylene, trimethylbenzene, tetramethylbenzene, ethyltoluene, diethylbenzene, triethylbenzene, chlorotoluene, dichlorotoluene, trichlorotoluene, chloroethylbenzene, dichlorobenzene, trichlorobenzene, tetrachlorobenzene, bromotoluene, bromoethylbenzene, dibromobenzene, dibromotoluene, chlorobromobenzene, dibromochlorobenzene, dichlorobromobenzene, etc. Of those, the method of the invention is especially favorable to the production of chlorotoluene, dichlorotoluene, chloroethylbenzene, dichlorobenzene and trichlorobenzene.

In the invention, a zeolite-containing adsorbent is used for adsorptive separation of aromatic compound isomers such as those mentioned above.

Zeolite for use herein is not limited to crystalline aluminosilicates only, but includes any others, for

example, crystalline aluminophosphates and silicoaluminophosphates such as ALPO, SAPO, etc..

The adsorbent shall be suitably selected, depending on the type of the compound to be separated with it. For example, for adsorptive separation of xylene, preferred is faujasite zeolite or the like; but for adsorptive separation of halogenoaromatic compounds such as chloroaromatic compounds, preferred is faujasite zeolite, pentasil zeolite or the like. These zeolite-containing adsorbents may be ion-exchanged with alkali metal salts, alkaline earth metal salts or the like for controlling their adsorption selectivity. For facilitating their use on an industrial scale, the adsorbents are preferably shaped along with a binder such as alumina, bentonite, etc.

The desorbent for use in the invention is not specifically defined. It includes, for example, aromatic compounds such as toluene, xylene, naphthalene, chlorobenzene, dichlorotoluene, etc. For separating heterocyclic compounds according to the invention, further usable are, in addition to the above-mentioned desorbents, any of aniline, methylaniline, dimethylaniline, diphenylamine, triphenylamine, toluidine, anisidine, chloroaniline, bromoaniline, nitroaniline, dinitroaniline, trinitroaniline, phenylenediamine, methylamine, dimethylamine, ethylamine, triethylamine, n-propylamine,

di-n-propylamine, tri-n-propylamine, isopropylamine, n-butylamine, isobutylamine, sec-butylamine, tert-butylamine, cyclohexylamine, benzylamine, α -phenylethylamine, β -phenylethylamine, ethylenediamine, tetramethylenediamine, hexamethylenediamine, piperazine, piperidine, pyrrolidine, etc.

In the invention, it is important to remove impurities that may be in the desorbent and will degrade the adsorbent being used, from the desorbent. Preferably, the impurity content of the desorbent to be circulated and reused in the invention is as small as possible. Concretely, it is desirable that the impurity content is at most 1000 ppm by weight, more preferably at most 100 ppm by weight. Removing impurities from the desorbent is effective for preventing the desorbent being used from being degraded and for ensuring efficient adsorptive separation of aromatic compound isomers.

The method of measuring the impurities in the desorbent varies, depending on the type of the impurities. For example, the impurities therein may be measured through gas chromatography, Karl Fischer's moistometry, Beckman's dissolved oxygen analysis, colorimetry under heat, etc.

For removing the impurities from it, for example, the desorbent is processed through distillation,

desorption, or adsorption to a solid adsorbent, or a part of the used desorbent is replaced with an impurity-free fresh desorbent.

Concretely, the desorbent that contains impurities is distilled to remove from it the impurities having a lower boiling point and/or the impurities having a higher boiling point through evaporation. The distillation may be effected in any ordinary manner. For example, the distillation tower to be used may be any of a plate column tower, a packed tower, etc., and the condition for operating it is not specifically defined.

For removing the impurities from the desorbent through desorption, for example, employable is any of a method of directly bubbling N_2 into the desorbent in a storage tank, or a method of contacting the desorbent with N_2 in countercurrent streams of the two in a plate column tower, a packed tower, etc. The condition for the methods is not specifically defined.

The desorbent may be processed through adsorption to a solid adsorbent for removing impurities from it. For example, the desorbent that contains impurities is contacted with a solid adsorbent so that the impurities are selectively adsorbed by the solid adsorbent, whereby the impurities are removed from the desorbent. For contacting the desorbent with a solid adsorbent,

employable is any of a fixed bed, a fluidized bed, etc. The solid adsorbent includes, for example, activated charcoal, zeolite, acid clay, activated clay, alumina, etc.

Of the impurities that may accumulate in the desorbent used in the process of adsorptive separation, highly polar substances such as water, hydrochloric acid, phenols and others strongly adsorb onto the surface of the adsorbent to lower the adsorptive separation capability of the adsorbent. Of those, dimers of aromatic compounds will be carbonated in the adsorbent to thereby also lower the adsorptive separation capability of the adsorbent. Therefore, removing these impurities from the desorbent, if possible, will be effective for preventing the adsorbent from being degraded, whereby the regeneration cycle of the adsorbent could be prolonged. Accordingly, in the invention, it is desirable to remove the impurities of water, hydrochloric acid, phenols, dimers of aromatic compounds, unsaturated hydrocarbon-containing compounds, and also oxygen-containing compounds such as those having aldehyde groups, carboxyl groups and the like, from the desorbent.

In the method of the invention, all or part of the desorbent to be circulated and reused in the adsorptive separation step may be processed in the previous step of removing the impurities from it, and it may be supplied to

the impurities removing step either continuously or intermittently.

In the method of the invention, the adsorptive separation may be effected in any known manner. For example, it may be effected in any of a fixed bed, a fluidized bed or a moving bed, in any manner of a flow process or batch process. The isomer mixture and the desorbent to be processed for such adsorptive separation may be in a vapor phase or a liquid phase. One typical process for adsorptive separation is a simulated moving bed process.

EXAMPLES

The invention is described concretely with reference to the following Examples, which, however, are not intended to restrict the scope of the invention.

Example 1:

Na-Y type zeolite having a molar ratio $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 4.8 (from Shokubai Kasei) is prepared according to the method described in the section of Examples in Japanese Patent Laid-Open No. 15833/1973, and ion-exchanged into an adsorbent of K-Y type zeolite. Using this, paraxylene (hereinafter referred to as PX) is separated from a mixture essentially comprising xylene isomers and ethylbenzene (this is a starting mixture to be subjected

to adsorptive separation herein, and is hereinafter referred to as a starting mixture). For the system to be used herein, the simulated moving bed system described in Example 5 in Japanese Patent Laid-Open No. 330025/1992 is so modified that it has 24 adsorption chambers. The outline of the system used herein is shown in Fig. 1; the condition for adsorptive separation in the system is in Table 1; and the composition of the starting mixture is in Table 2. In this system, used is at least 90 wt.% paradiethylbenzene (hereinafter referred to as p-DEB) as the desorbent, and the starting mixture is subjected to adsorptive separation. The intended product, PX is taken out as an extract flow along with the desorbent; and the remaining PX not having been recovered in the extract flow and the other components of the starting mixture are taken out as a raffinate flow along with the desorbent. The extract and the raffinate are then processed in the distillation units 1 and 2, respectively, to recover the desorbent. The thus-recovered desorbent is circulated and reused in the system along with the desorbent also recovered from the 24th adsorption chamber. When contacted with the adsorbent at high temperatures in the system, the starting mixture and the desorbent will form high-boiling-point substances such as dimers, etc.; or when contacted with minor O₂ in the N₂ streams in the

distillation units, they will form oxygen-containing compounds having an aldehyde group or a carboxyl group. Though their amount is small, these impurities will accumulate in the desorbent to be circulated and reused in the system whereby the adsorptive separation capability of the system is lowered.

As an index of the separation capability of the system, employed is the "adsorption recovery" for 99.80 wt.% product purity to be defined by the following equation, and its time-dependent change is plotted, based on the adsorption recovery at the start of the system.

Adsorption Recovery

= [(amount of PX recovered as product, g/hr)/(amount of PX in Xylene isomer mixture processed for adsorptive separation, g/hr)] \times 100

In this Example, a part of the desorbent is continuously extracted out and processed in the distillation unit 3 to remove the high-boiling-point substances from it, and then circulated and reused in the system. Accordingly, the adsorption capability of the system is not lowered as in Fig. 2. On the other hand, the high-boiling-point substances having been removed from the desorbent are concentrated and analyzed through gas chromatography, for which the condition is shown in Table 3. From the chart of the gas chromatogram shown in Fig. 3,

it is seen that many high-boiling-point substances are in the desorbent.

Comparative Example 1:

The same absorptive separation process as in Example 1 is repeated herein. In this, however, the step of continuously extracting a part of the desorbent to be circulated and reused in the system, and processing it for removing impurities therefrom through distillation is omitted. The adsorption recovery in the system is greatly lowered, as in Fig. 2.

Comparative Example 2:

First, the adsorptive separation process of Comparative Example 1 is repeated for a while, but in the halfway through it, this is combined with the step of desorbent purification as in Example 1. In this, the adsorptive recovery in the system is greatly lowered before the halfway, but the reduction in the adsorptive recovery is stopped after the start of the step of removing impurities, as in Fig. 2.

Example 2:

A zeolite adsorbent for separation of m-dichlorobenzene (hereinafter referred to as m-DCB) is prepared according to the method described in Examples 1 to 4 in Japanese Patent Laid-Open No. 330025/1992. This is Y-type zeolite containing cations of potassium and lead

ions. The adsorbent is baked at 500°C for 2 hours, and set in a simulated moving bed system for adsorptive separation, as in Example 5 in Japanese Patent Laid-Open No. 330025/1992. The outline of the system used herein is shown in Fig. 4; and the condition for adsorptive separation in the system is in Table 4. In this system, used is 3,4-dichlorotoluene (hereinafter referred to as 3,4-DCT) is used as the desorbent, and a DCB isomer mixture is subjected to adsorptive separation. The intended product, m-DCB is taken out as a raffinate flow along with the desorbent; and the remaining m-DCB not having been recovered in the raffinate flow and the other components o-DCB and p-DCB are taken out as an extract flow along with the desorbent. The extract and the raffinate are then processed in the distillation units 1 and 2, respectively, to recover the desorbent. The thus-recovered desorbent is circulated and reused in the system along with the desorbent also recovered from the adsorption chamber 12. When heated, 3,4-DCT forms a small amount of high-boiling-point substances such as biphenylmethanes, etc. The impurities generally accumulate in the desorbent to be circulated and reused in the system whereby the adsorptive separation capability of the system is lowered.

As an index of the separation capability of the

system, employed is the adsorption recovery for 99.5 % product purity to be defined by the following equation, and its time-dependent change is plotted, based on the adsorption recovery at the start of the system.

Adsorption Recovery

$$= \left[\frac{\text{(amount of m-DCB recovered as product, g/hr)}}{\text{(amount of m-DCB in DCB isomer mixture processed for adsorptive separation, g/hr)}} \right] \times 100$$

In this Example, a part of the desorbent is continuously extracted out and processed in the distillation units 3 and 4 to remove the impurities from it, and then circulated and reused in the system. Accordingly, the adsorption capability of the system is not lowered as in Fig. 5. On the other hand, the high-boiling-point substances having been removed from the desorbent are concentrated and analyzed through gas chromatography, for which the condition is shown in Table 5. From the chart of the gas chromatogram shown in Fig. 6, it is seen that many high-boiling-point substances are in the desorbent.

Table 1

Item	Condition
Adsorption Chambers	16 ml × 24 chambers
Amount of Desorbent fed into system	138.7 gr/hr
Amount of Starting Mixture fed into system	62.7 gr/hr
Amount of Extract Flow	61.6 gr/hr
Amount of Raffinate Flow	139.8 gr/hr
Adsorption Temperature	175°C
Amount of Desorbent Flow Circulated and Reused after Distillation	1.6 gr/hr

Table 2

Component	Concentration
Ethylbenzene	7.68 wt. %
Paraxylene	21.23 wt. %
Metaxylene	49.19 wt. %
Orthoxylene	21.90 wt. %

Table 3

Item	Condition
Detector	FID
Column	Thermol-3 (5 %), Celite 545
Column Temperature	100°C → 240°C (programmed temperature gas chromatography)
Inlet Port Temperature	250°C
Heating Rate	8°C/min
Time at which heating started	240 seconds after sample introduction
Carrier Gas	N ₂ , 30 cc/min
Amount of Sample applied to column	1 μ l

Table 4

Item	Condition
Capacity of Adsorption Chambers	16 ml \times 12 chambers
Amount of Desorbent fed into system	364 ml/hr
Amount of DCB Isomer Mixture fed into system	16 ml/hr
Amount of Extract Flow	71 ml/hr
Amount of Raffinate Flow	37 ml/hr
Adsorption Temperature	130°C
Time for switching simulated moving beds	about 150 seconds
Amount of Desorbent Flow Circulated and Reused after Distillation	12 ml/hr

Table 5

Item	Condition
Detector	FID
Column	Silicon OV-17 (5 %)/Unipor B, 60-80 mesh, 2 m \times 3 mm ϕ
Column Temperature	250°C
Inlet Port Temperature	300°C
Carrier Gas	N ₂ , 225 kPa
Air	50 kPa
Hydrogen	60 kPa
Amount of Sample applied to column	1 μ l

Comparative Example 3:

The same absorptive separation process as in Example 2 is repeated herein. In this, however, the step of continuously extracting a part of the desorbent to be

circulated and reused in the system, and processing it for removing impurities therefrom through distillation is omitted. The adsorption recovery in the system is greatly lowered, as in Fig. 5.

Comparative Example 4:

First, the adsorptive separation process of Comparative Example 1 is repeated for a while, but in the halfway through it, this is combined with the step of desorbent purification as in Example 2. In this, the adsorptive recovery in the system is greatly lowered before the halfway, but the reduction in the adsorptive recovery is stopped after the start of the step of removing impurities, as in Fig. 5.

Example 3:

To 100 parts by weight of sodium-type Y-type zeolite (hereinafter referred to as Na-Y) (powdered product of Zeolum Na-5.1Y from Toso), added is 15 parts by weight, in terms of alumina, of alumina sol (Nissan Chemical's Alumina Sol #200 with $\text{Al}_2\text{O}_3 = 10$ wt.%) serving as a binder, and granulated into 0.15 to 0.5 mm ϕ granules. The NaY-type zeolite granules are dried at 120°C, and then baked at 500°C for 2 hours. With an aqueous solution of silver nitrate that corresponds to 50 % (in terms of the metal by mol) of the Na cation site of Na-Y, the zeolite granules are ion-exchanged (solid/liquid ratio, 3.0 liters/kg) by

keeping them therein at room temperature for 30 minutes and then heating them at 85°C for 1 hour. These are then fully washed with distilled water, dried at 120 °C, and then baked at 500°C for 2 hours.

Using the adsorbent (Ag-Na-Y) together with 2,4-xylydine serving as a desorbent, a mixture of α -picoline: β -picoline:2,6-lutidine:2,4-lutidine = 1:1:1:1 (by weight) is processed for adsorptive separation in the same manner as in Example 2.

The intended product, 2,6-lutidine is taken out as a raffinate flow along with the desorbent; and the remaining 2,6-lutidine not having been recovered in the raffinate flow and the other components α -picoline, β -picoline and 2,4-lutidine are taken out as an extract flow along with the desorbent.

As an index of the separation capability of the system, employed is the adsorption recovery for 99.5 % product purity to be defined by the following equation, and its time-dependent change is plotted, based on the adsorption recovery at the start of the system.

Adsorption Recovery

$$= \left[\frac{\text{(amount of 2,6-lutidine recovered as product, g/hr)}}{\text{(amount of 2,6-lutidine in substituted pyridine mixture processed for adsorptive separation, g/hr)}} \right] \times 100$$

In this Example, a part of the desorbent is

continuously extracted out and processed in the distillation units 3 and 4 to remove the impurities from it, and then circulated and reused in the system. Accordingly, the adsorption capability of the system is not lowered as in Fig. 7.

Comparative Example 5:

The same absorptive separation process as in Example 3 is repeated herein. In this, however, the step of continuously extracting a part of the desorbent to be circulated and reused in the system, and processing it for removing impurities therefrom through distillation is omitted. The adsorption recovery in the system is greatly lowered, as in Fig. 7.

While the invention has been described in detail and with reference to specific embodiments thereof, it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

Claims:

1. A method for producing an aromatic compound isomer substituted with alkyl group(s) and/or halogen atom(s), through adsorptive separation by the use of a zeolite-containing adsorbent and a desorbent, wherein the desorbent is, after having been processed for removing impurities from it, supplied to the adsorptive separation step.

2. The method for producing an aromatic compound isomer as claimed in claim 1, wherein the aromatic compound has a benzene ring or heterocyclic ring structure.

3. The method for producing an aromatic compound isomer as claimed in claim 1 or 2, wherein the aromatic compound is ring-substituted with at least one halogen element.

4. The method for producing an aromatic compound isomer as claimed in any of claims 1 to 3, wherein the impurities in the desorbent are removed through any of distillation, purging or adsorption to a solid adsorbent.

5. The method for producing an aromatic compound isomer as claimed in any of claims 1 to 3, wherein the impurities in the desorbent are removed by replacing a part of the used desorbent with an impurity-free fresh desorbent.

6. The method for producing an aromatic compound

isomer as claimed in any of claims 1 to 5, wherein all or part of the desorbent to be supplied to the adsorptive separation step is first continuously or intermittently supplied to a step of removing impurities from it, and then supplied to the adsorptive separation step.

ABSTRACT OF THE DISCLOSURE

In a process of producing an aromatic compound isomer substituted with alkyl group(s) and/or halogen atom(s), through adsorptive separation by the use of a zeolite-containing adsorbent and a desorbent, the desorbent is, after having been processed for removing impurities from it, supplied to the adsorptive separation step. In the process, the adsorbent used is prevented from being degraded, and its life is prolonged to efficiently separate an aromatic compound isomer mixture into individual isomers.

Fig. 1

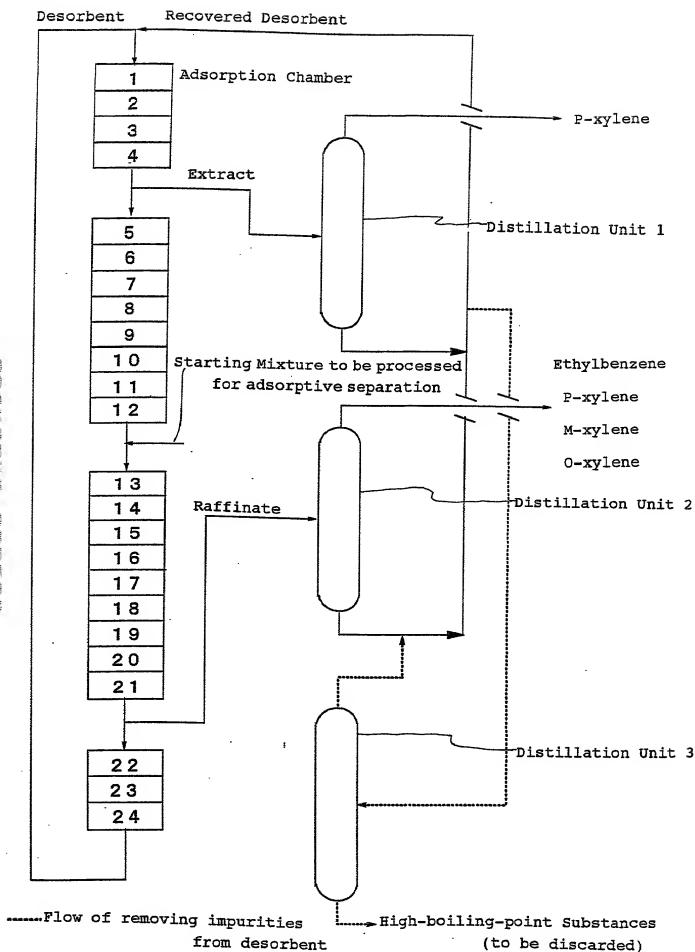


Fig. 2

Adsorption Recovery (wt.%)

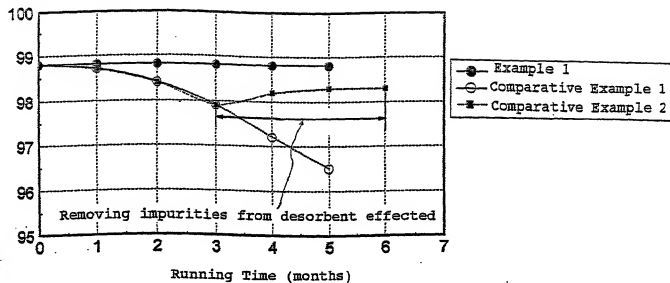


Fig. 3

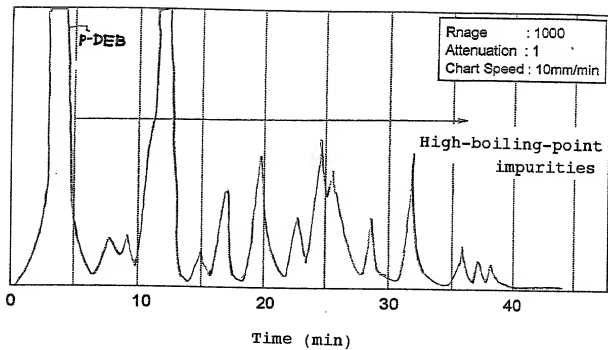


Fig. 4

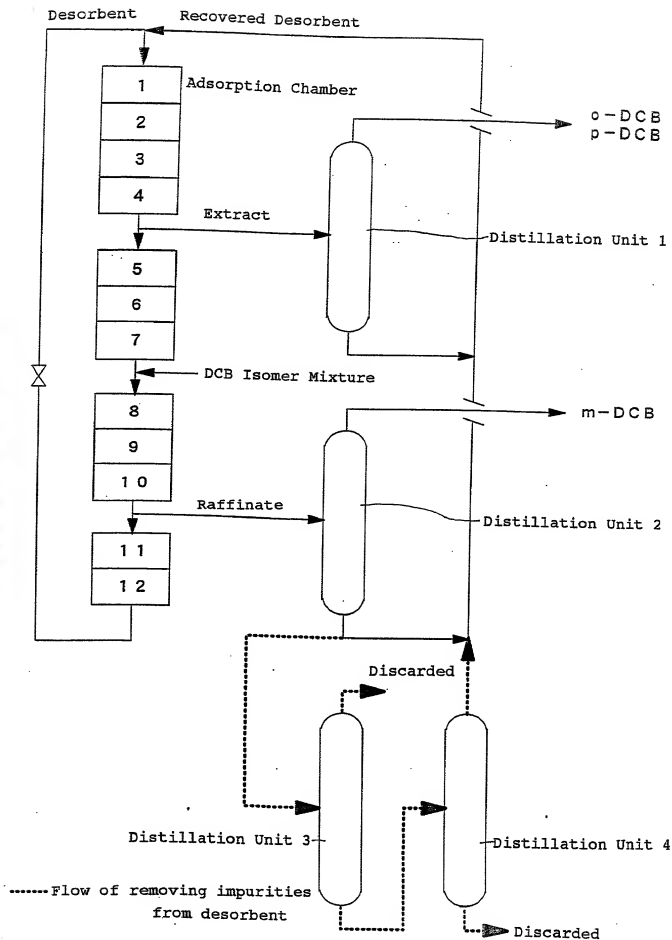


Fig. 5

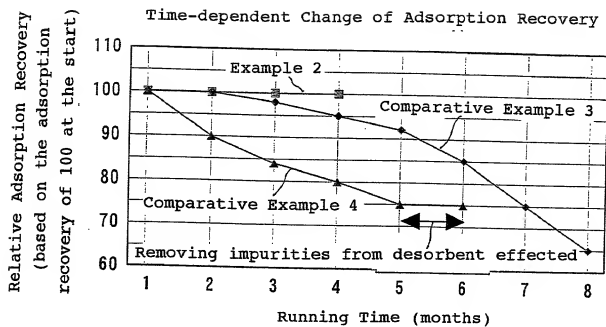


Fig. 6

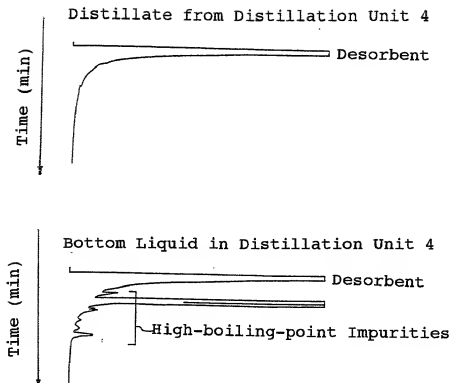
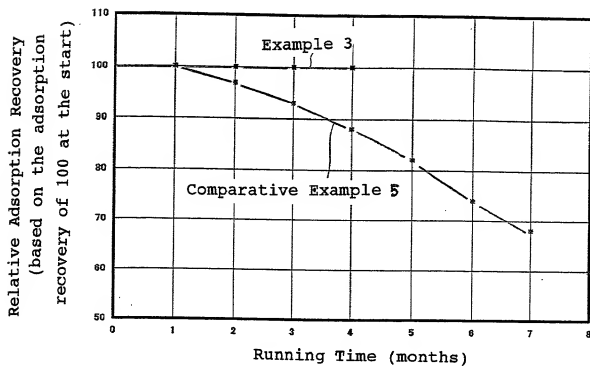


Fig. 7 Time-dependent Change of Adsorption Recovery



- ☒ Original Application
☐ PCT National Application
U.S. Designated Office
☐ Continuation or Divisional Application
☐ Continuation-in-Part Application

**COMBINED DECLARATION,
POWER OF ATTORNEY AND PETITION**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled METHOD FOR PRODUCING AROMATIC COMPOUND ISOMERS

- ☒ which is described in the specification and claims

☒ attached hereto.

☐ filed on _____

Application Serial No. _____

and was amended on _____

(if applicable)

- ☐ which is described in International Application No. _____

filed _____ and as amended on _____

_____ (if any),
which I have reviewed and for which I solicit a United States patent.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I do not know and do not believe that this invention was ever known or used in the United States before my or our invention thereof or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application or said international application, or in public use or on sale in the United States of America more than one year prior to this application or said international application, or that the invention has been patented or made the subject of an inventor's certificate issued before the date of this application or said international application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months prior to this application or said international application, or that any application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application or said international application by me or my legal representatives or assigns except as identified below.

COMBINED DECLARATION, POWER OF ATTORNEY AND PETITION

(Page 2)

Attorney Docket No. 1382-00

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International Application which designated at least one country other than the United States of America, listed below and have also identified below any foreign application(s) for patent or inventor's certificate, or of any PCT International Application having a filing date before that of the application on which priority is claimed:

Number	Country	Date of Filing (day,month,year)	Priority Claimed
H11-240393	Japan	August 26, 1999	<input checked="" type="checkbox"/> yes <input type="checkbox"/> no
			<input type="checkbox"/> yes <input type="checkbox"/> no
			<input type="checkbox"/> yes <input type="checkbox"/> no
			<input type="checkbox"/> yes <input type="checkbox"/> no
			<input type="checkbox"/> yes <input type="checkbox"/> no

I hereby claim the benefit under Title 35, United States Code, §119(e) or §120 (as applicable) of any United States application(s), or §365(c) of any PCT International Application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International Application(s) in the manner provided by the first paragraph of Title 35, United States Code, §112:

(Application Serial No.)

(Filing Date)

(Status)(patented,pending,abandoned)

(Application Serial No.)

(Filing Date)

(Status)(patented,pending,abandoned)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the registered attorneys listed under Customer No. 022469 and the following registered attorneys to prosecute this application and transact all business in the United States Patent and Trademark Office connected therewith:

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(Page 3)

Attorney Docket No. 1382-00

I hereby petition for grant of a United States Letters Patent on this invention.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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POST OFFICE ADDRESS			
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